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OZONE IN WATER TREATMENT

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PERSPECTIVE OF COMBINATION OF OZONE AND ULTRASOUND

INTRODUCTION: In the field of water works, the advantage of a combination of two agents to obtain more favourable action, than if these were used separately, is obvious. By itself, water is a poor cleaner, soap is not a cleaner at all, but the combination of the two provides a convenient, efficacious, rapid and economical cleansing. Yet disinfection of water is based on the separate use of few single agents. These "poor cleaners" have been investigated and utilized for a period of 100 years, but the beneficial "soaps" remain to be researched and developed.

This chapter deals with one of the potential combinations: SONOZONE.

SYNERGISTIC EFFECTS: In gas/liquid semiflow laboratory experiments, ultrasonic treatment, 10 W/ℓ, 20 kHz, was able to increase the rate constants of ozone oxidation of rhodamine-B by 37-55%, figure 1. Similar synergisms were reported for reduction of Chemical Oxygen Demand and Total Organic Carbon (6). The liquid/liquid technique and the non-simultaneous combination of ultrasound and gaseous ozone are not capable of reproducing these effects.

The employed ultrasonic treatment in itself, does not cause significant inactivation of virus and bacteria. On the contrary, doubling of the microbial count has been reported. Nevertheless, pre- as well as simultaneous sonication are able to bring about a synergistic action with ozone in the gas/liquid- and, to less extent, in liquid/liquid techniques, see table 1. Seventy per cent of the ozone dosage required for 3 decades inactivation, and eighty per cent of the ozone dosage required for 4 decades inactivation by gaseous ozone alone, could be saved by the simultaneous application of ultrasound in steady state laboratory experiments (5), see figure 2.

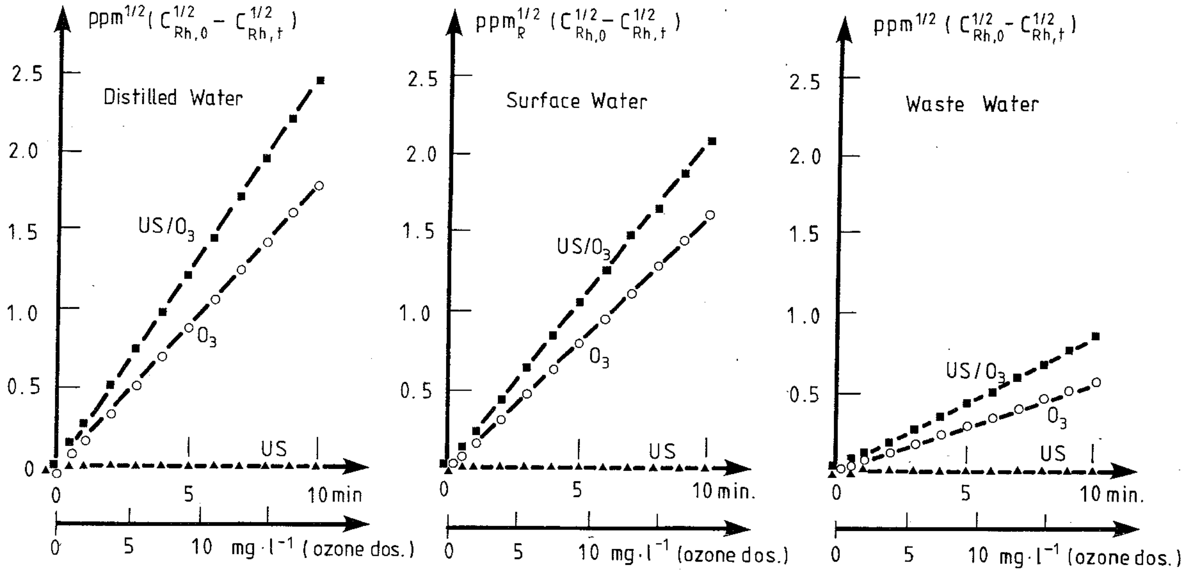

Figure 1. Decolorization of Rhodamine-B vs. time with ultrasonic treatment, with gaseous ozone dosage, and with simultaneous combination of the two in semiflow experiments (2).

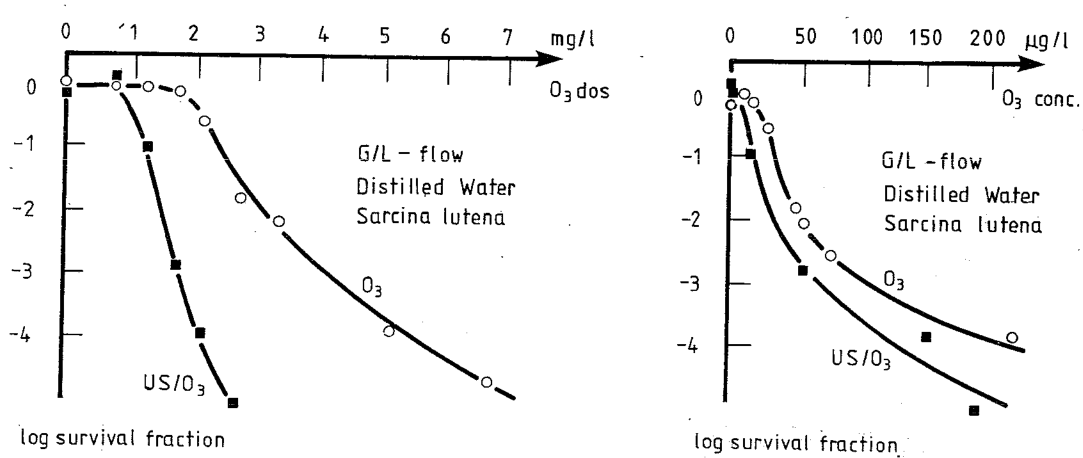

Figure 2. Steady state inactivation of bacteria by gaseous ozone with and without ultrasonic treatment (5).

Thus, in combination the effect of ultrasound and ozone is synergistic chemically as well as microbiologically. Under certain circumstances however, the combination effects may be additive or even antagonistic.

INTERACTIONS AND MECHANISMS: The pattern of interactions between ozone and ultrasound is very complex, and not completely understood. In the ozonation processes, gaseous ozone is transferred to the water and the dissolved ozone reacts with the contents of dissolved chemicals, suspended solids and microorganisms. Dissolved ozone also produces free radicals, some of which are able to initiate chain reactions with ozone and to oxidize dissolved chemicals and probably to inactivate microorganisms, figure 3.

Propagation of intensive ultrasonic waves in the water, brings out the interference phenomenon called cavitation. Cavitational ultrasound is able to act on water and, via a free radical mechanism, to produce peroxide at µg/ℓ levels. It is also capable of breaking chemical bonds and of transforming small amounts of a wide variety of dissolved chemicals.

Cavitational ultrasonic irradiation of dissolved ozone increases the ozone autodecomposition significantly, presumably by back-feeding its chain transfer to free radicals. Furthermore, the direct molecular ozone reactions may be accelerated due to the local heat and pressure effects of cavitation. The vibration energy is, in itself, not great enough to cause chemical changes.

Ultrasound acts on ozone bubbles and increases the mass transfer between gas and the liquid. This effect has been explained as a matter of disruption of the ozone bubbles and of expansion of the gas/liquid contact surface. Indeed, small bubbles often increase in size to resonate. At the resonance, the microscopic turbulence of the cavitational and/or vibratory energy increase the gas/liquid mass transfer, apparently even though the macroscopic turbulence of some mixing device is already high.

The action of ultrasonic irradiation is reported to have several effects on microorganisms in water. Both growth and inactivation

Table 1. Division and characteristics of the function of inactivation by ozone with and without ultrasound.

Curve segment	Characteristic	Suggested nomenclature	Explanation
Stage 1	Almost horizontal	Ozone demand stage	Prolonged by addition of ozone consuming pollutants and by presonication of polluted water. Reduced by simultaneous sonication in gas/liquid techniques.
Stage 2	Steep slope, deflection tangent	Proper inactivation stage	Apparently not affected by ultrasound. Decreased slope in flow technique and by very slow ozone dosages in semiflow technique.
Stage 3	Slow slope	Aggregate inactivation stage	Eliminated or reduced by sonication and by filtration prior to ozonation.

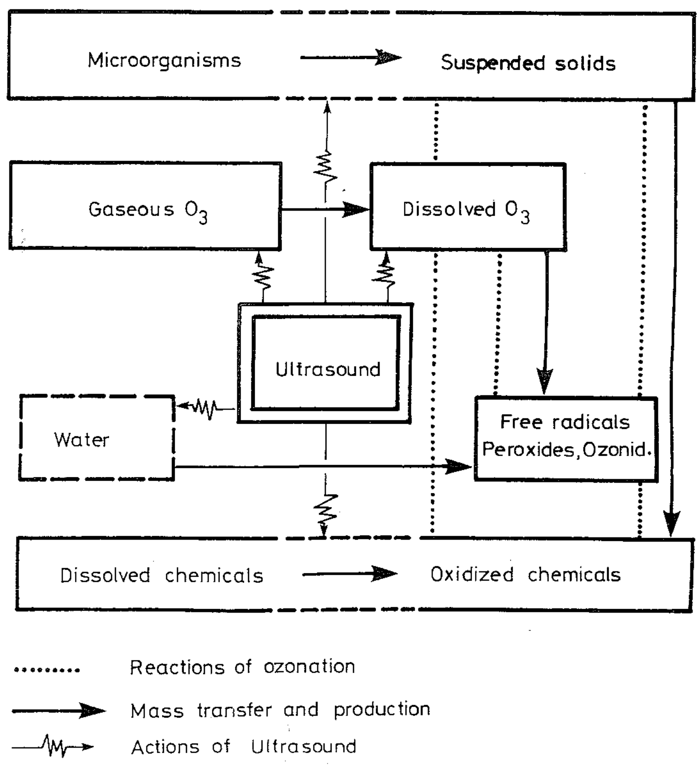

Figure 3. Simplified schematic diagram of the processes and interactions of sonozone.

may take place, depending upon, among other things, the ultrasonic intensity and the duration of the treatment. At the intensities and residence times, which may be realistic for water treatment, the disaggregation of microbial flocs is of major importance.

Ozone is very efficacious and rapid to perform the first few decades of inactivation, but is inert and slow to inactivate the residual microbial flocs, which is encapsulated. Surviving bacteria in flocs may be protected by refractive organic matter, which is made biodegradable by ozone itself (1,3). Thus, at the time where ozone has disappeared, the encapsulated infections material will be vital and cause aftergrowth. For this reason sonozone, by disaggregation of flocs while ozone is still present, may give a more efficient inactivation and reduce the aftergrowth problems of water works.

METHODS AND TECHNOLOGY: Our knowledge on the methods and technology of sonozone is extremely limited. No results have been reported on sonozonation of authentic untreated or partly treated drinking waters; neither in laboratory-, nor in pilot plant equipment. Indeed, the techniques employed have been chosen more or less blindly. The sonozonation technology of today, is to be compared with the ozonation technology of the beginning of this century.

Ultrasound is usually produced by piezoelectric transducers connected with a generator. The anisotropic crystals of the transducers, f.ex. barium titanate, are exposed to a reversing electric field. This makes the crystals expand and contract periodically, and generates the acoustic waves in the adjacent medium.

It is evident that ultrasound and ozone can be combined in many different ways. Presumably, the simultaneous treatment in directly injected ozonation shafts is the most promising method, figure 4. Even though most experiences have been gained at 20 kHz, ultrasound is believed to act synergistic at a very wide range of frequencies. The intensity is supposed to be of greater importance. Netto intensities of the order 1 W/cm^2 have been employed successfully in the laboratory experiments. Apparently, maximum effects would be obtained if the cavitational ultrasonic waves are focused closely over the site of initial contact between ozone bubbles and the flowing water. In such a

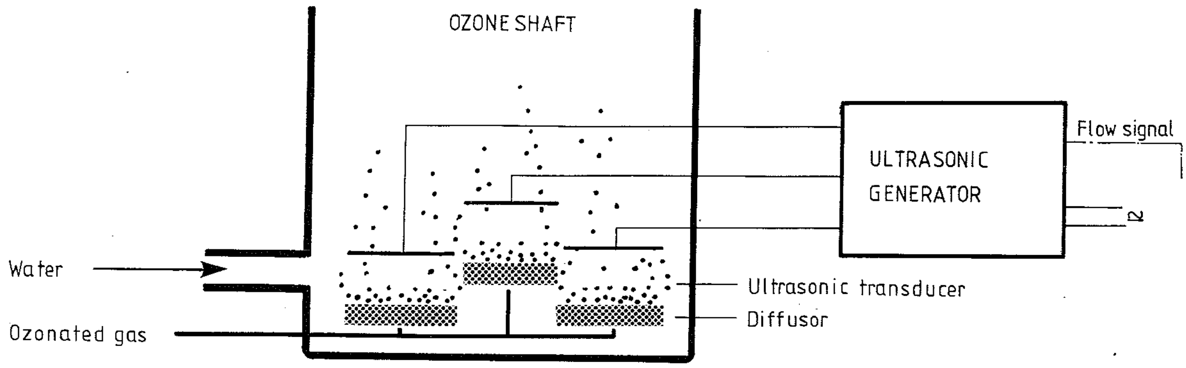

Figure 4. Suggested sonozone equipment.

system the water flow can be increased beyond what has until now been attempted.

ECONOMICS AND FURTHER DEVELOPMENTS: At waterworks dose levels, the effects of ultrasound have been valuated as follows: if the ozone dose D_{O_3} is required to obtain a given effect by ozonation, and a smaller ozone dose d_{O_3} is required by an equal sonozonation at which the ultrasound dose d_{US} is employed, then:

$$UE_{sonozone} = 2.6\,[(1-SF)\cdot D_{O_3}]^{0.5} + 0{,}14\, d_{US}^{0.9}$$

where: UE is the uniform expense in Danish Øre/m^3
D,d is the dose in mg O_3/ℓ or Wh ultrasound /m^3
SF is the ozone saving factor = $1 - d_{O_3}/D_{O_3}$.

A sonozonation with assumed 50 Wh/m^3 ultrasonic dosage and 0.7 saving factor, was estimated to be slightly more expensive than ozonation alone (4). Thus, the effective synergism observed seems, at the moment and from an economical point of view, not to be attractive for waterworks.

Of course, this balance sheet would easily be turned around, if the laboratory results cannot be reproduced at a technical scale. On the other hand, technical development by optimization of the employed ultrasonic intensity and frequency or by production of less expensive and more durable transducers, would turn the balance sheet the other way around.

However, toxicological, microbiological, chemical or technical aspects, about which we still know very litte, may be of greater importance than the economic ones for the choice of disinfection methods at waterworks.

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